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Contact Angle of Yucca Mountain Welded Tuff with Water and Brines

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ABSTRACT

A number of tests were performed to acquire contact angles between Yucca Mountain welded tuff from Topopah Springs Lower Lithophysal geologic unit and various brine solutions. The tests were performed on core disks received from Sample Management Facility (SMF), oven dried to a constant weight and the core disks vacuum saturated in: distilled water, J-13 water, calcium chloride brine and sodium chloride brine to constant weight. The contact angles were acquired from eight points on the surface of the core disks, four on rough surface, and four on polished surface. The contact angle was measured by placing a droplet of the test fluid, distilled water, J-13 water, calcium chloride brine and sodium chloride brine on the core disks. The objective of this test was to acquire contact angles as a potential input to estimating capillary forces in accumulated dust on the waste packages and drip shields slated for the proposed High-Level Radioactive Waste Repository at Yucca Mountain, Nevada. It was noted that once the droplet contacts the test surface, it continues to spread hence the contact angle continues to decrease with elapsed time. The maximum observed angle was at time 0 or when the drop contacted the rock surface. The measured contact angle, in all cases has significant scatter. In general, the time zero contact angles for core disks saturated in sodium chloride brine were smaller than those saturated in calcium chloride brine, distilled water, and J-13 water. The contact angles for samples saturated in distilled water, J-13 water and calcium chloride brine at time zero were similar. There was slight difference between the observed contact angles for smooth and rough surface of the test samples. The contact angles for smooth surfaces were smaller than for the rough surfaces.

I. INTRODUCTION

When a fluid drop is placed on a solid surface a liquid film is formed as it spreads out. The spread of the fluid is impended by the retarding velocity due to the influence of capillarity, surface tension and the tendency of some liquids to wet the surfaces. The spreading liquid film attains maximum spread when it comes to rest. The liquid film forms an angle while in contact with the solid surface. This angle is defined as contact angle and is generally referred to as wettability of the surface. The liquid is considered non-wetting at angles between $90^0<\theta<180^0$ and wetting when the angle is between $0^0<\theta<90^0$ complete wetting would be $\theta=0^0$. For this test, contact angles were measured between core disks from Yucca Mountain welded tuff Topopah Springs Lower Lithophysal geologic unit and various aqueous solutions expected at Yucca Mountain. The data would then be analyzed to ascertain inputs for estimating capillary forces in accumulated dust on the waste packages and drip shields slated for the proposed repository (1). Yucca Mountain is approximately 100 km north of Las Vegas and is the site for a proposed High-Level Radioactive Waste (HLW) from commercial and defense nuclear reactors. This paper describes the test configuration, data collected and analyzed to derive surface tension or capillarity between the tuff core disks and the aqueous solutions. .

II. WORK DESCRIPTION

This test consisted of preparing ten core disks, approximately 60 mm in diameter and 13 mm in thickness. It was realized from the onset that the surface of the core samples, as produced by saw cut, might not be uniform as compared to the polished metallic surfaces generally used for this type of measurement. Therefore, it was decided to medium polish one surface of the sample and keep one surface irregular as produced by the rock saw. The medium polished surface of the core was prepared in two steps. In step one, the sample surface was polished on a 30-micron grit wheel and then the sample was polished on 220-grit wheel. This resulted in one surface being relatively smooth and polished. . It should be noted that the surface of the samples, even the polished surface, were still microscopically quite rough. To account for the surface variability as well as the non-homogenous nature of the test samples, the contact angles were measured at four points on each side of the disk for a total of eight measurements. The drops were placed at nominally 0^0, 90^0, 180^0 and 270^0 with a clockwise orientation.

Table 1. Sample Environment[1]

Sample #	Sample Interval-m	Treatment
01026607	866.4-866.5	Saturated with calcium chloride
01026602	842.5-842.6	Saturated with calcium chloride
01026597	785.5-785.6	Saturated with calcium chloride
01026608	906.1-906.2	Saturated with sodium chloride
01026609	906.1-906.2	Saturated with sodium chloride
01026610	906.1-906.2	Saturated with sodium chloride
01026605	866.2-866.3	Saturated with distilled water
01026596	785.5-785.6	Saturated with distilled water
01026612	906.3-906.4	Saturated with J-13 water
01026606	866.3-866.4	Saturated with J-13 water

1. Parent borehole UE-25, and UZ#16

The contact angles were measured by placing a droplet of distilled water, J-13 water from Yucca Mountain, sodium chloride brine and calcium chloride brine. Once the droplet contacts the test surface, it continues to spread creating a changing fluid volume and contact angle. The fluid volume would change as the liquid was absorbed into the sample. The change in contact angle and volume appears to be affected by the micro features in the rock samples.

III. TEST APPARATUS

The contact angles were measured by a contact angle measuring apparatus manufactured by First Ten Angstrom, Model FTA 188 (2). It is a general-purpose laboratory instrument for measuring contact angles, surface tension and indirectly surface energy and fluid adhesion characteristics. The instrument used "drop shape" analysis method to make measurements. The contact angles are obtained by fitting a mathematical expression to the shape of the drop and then calculating the slope of the tangent to the drop at the liquid, solid, vapor interface. For the measurements reported in this paper, the FTA 188 came preloaded with software, FTA 32, version 2.0. This software automatically calculated the contact angle and other parameters such as base volume of the droplet and the surface tension.

The apparatus can be operated in video or movie mode. In video mode a single shot is captured for the droplet, whereas in movie mode the droplet characteristics are recorded over time. For this activity the contact angles were measured by setting the instrument in a movie mode rather than in video mode. The movie mode was selected to permit capturing the changes in the shape of the drop as the fluid film spread on the sample surface. Movie mode allowed for recording the change in drop shape, hence the contact angle over time as it dispersed on the sample surface or absorbed in the rock matrix. Thus, several contact angles were acquired for a given drop and test environment corresponding to elapsed time of 20 to 30 seconds. It should be noted that, in some tests the fluid dispersed within four to six seconds.

IV. TEST METHODOLOGY

The sodium chloride brine was prepared by dissolving 325 gm of reagent grade sodium chloride in 700 ml of water. The calcium chloride brine was prepared by dissolving 633 gm of reagent grade calcium chloride in 700 ml of distilled water. All solutions were prepared at room temperature. The contact angles were measured at room temperature as follows:

1. Measure contact angle by placing distilled water droplets on rough and polished surface of the sample as received.
2. Measure contact angle by placing J-13 water droplets from Yucca Mountain droplets on rough and polished surface of the sample as received.
3. Oven dry all samples to a constant weight.
4. Measure contact angle for some of the oven dry samples by placing distilled water droplets on rough and polished surface of the samples.
5. Measure contact angle by placing J-13 water droplets from Yucca Mountain droplets on rough and polished surface of the oven dry samples.
6. Vacuum saturate three samples in sodium chloride brine, three samples calcium chloride brine, two samples in distilled water and two samples in J-13 water.
7. Measure contact angle by placing droplets of distilled water on rough and polished surface of the sample vacuum saturated in distilled water.
8. Measure contact angle by placing droplets of J-13 water on rough and polished surface of the sample vacuum saturated in J-13 water.
9. Measure contact angle by placing droplets of saturated sodium chloride brine on rough and polished surface of the sample vacuum saturated in saturated sodium chloride brine
10. Measure contact angle by placing droplets of saturated calcium chloride brine on rough and polished surface of the sample vacuum saturated in saturated calcium chloride brine

Figure 1 shows a typical contact angle measurement for a sample saturated in calcium chloride brine with a calcium chloride brine droplet, a sample saturated in sodium chloride brine with sodium chloride brine droplet, a sample saturated in distilled water with a distilled water droplet and the test apparatus performance check performed using a 90^0 standard.

V. TEST RESULTS

Although, the test samples were from the same parent core, the measured contact angles were different for each test sample for a given droplet location. The test results for all saturated samples are found in Figures 2 and 3. The bar charts in Figure 2 and 3 represents contact angles measured at eight locations. Figures 4 and 5 are average contact angles plotted against the elapsed time. The elapsed time is in seconds after the droplet is placed on the surface of the sample. The average angle is the average of the four measurements taken at 0^0, 90^0, 180^0 and 270^0 along the surface. The measurements were repeated for both the rough and semi-polished surfaces of the test samples for eight measurements. The plots represent change in contact angle as the fluid spreads on the test sample. Table 2 is a summary of contact angles measured for saturated and unsaturated samples. The key observations from Table 2 are:

1. The average contact angle for the two unsaturated test samples consisting of eight measurements, using distilled water and J-13 water, were 39.20^0 and 47.98^0 for the polished surface. The average contact angles for rough surface for the samples with distilled water and J-13 water were 50.10^0 and 59.35^0 respectively.
2. The average contact angle for samples saturated with distilled water or J-13 water was determined from eight measurements each for the polished and rough surfaces of the two samples. For the polished surface using distilled water or J-13 water droplets, the contact angles were 65.63^0 and 61.96^0 respectively. For the rough surface for the same samples again using distilled water J-13 water droplets, the contact angle were 75.89^0 and 64.570 respectively.
3. The average contact angle for three samples saturated with sodium chloride brine with sodium chloride brine droplets, was 12.41^0 for polished surface and 13.94^0 for the rough surface.

Table 2A. Summary contact angle for polished surface

Sample Env.	Avg. deg.	Max deg.	Min deg.	Std D deg.	Test Fluid
Sat Nacl	12.41	23.60	5.77	4.82	Na brine
Sat. $Cacl_2$	37.89	71.99	27.25	14.79	Ca. brine
Sat DW	65.63	81.90	47.89	10.87	DW
Sat J-13	61.96	85.84	51.75	10.97	J-13
UnSat-DW	39.20	60.21	23.11	14.10	DW
Unsat. J-13	47.98	68.47	27.32	16.01	J-13

Table 2B. Summary contact angle for rough surface

Sample Env.	Avg. deg	Max deg.	Min deg.	Std D deg.	Test Fluid
Sat Nacl	13.94	21.23	8.26	3.47	Na brine
Sat. $Cacl_2$	48.84	83.40	26.13	17.87	Ca. brine
Sat DW	75.89	87.31	54.38	10.91	DW
Sat J-13	64.57	85.84	60.85	11.72	J-13
UnSat-DW	50.10	65.09	21.21	14.82	DW
Unsat J-13	59.35	84.71	27.32	16.01	J-13

Note:

Avg. Average
DW: distilled water
Env. Environment
Sat. Saturated
UnSat. Unsaturated

4. The average contact angle for three samples saturated with calcium chloride brine with calcium chloride brine droplets was 37.89^0 for the polished surface and 48.84^0 for the rough surface.

VI. APPLICATION OF THE TEST RESULTS

The primary objective of this test was to acquire contact angles as a potential input for estimating capillary forces in accumulated dust on the waste packages and drip shields slated for the proposed repository (1). This objective has been accomplished. With the exception of samples saturated in sodium chloride brine with sodium chloride brine droplets, the acquired data can be used to estimate capillary forces on accumulated dusts. The measured angles are highly dependent upon the microscopic characteristics of the test sample. For the sodium chloride brine samples, the standard deviation varies significantly making it difficult to distinguish a correlation necessary for estimating the capillary forces.

VII. SUMMARY

The test methodology and equipment performed well. Ten samples from Yucca Mountain welded tuff Topopah Springs Lower Lithophysal geologic unit were used to measure contact angles under a varying sample environment.

1. It is difficult to acquire precise measurements with a limited number of test samples. To achieve higher level confidence in the data a large number of tests should be performed with solution of different concentration.
2. The contact angles for distilled water with sample saturated in distilled water were much larger than the samples saturated in sodium chloride brine or calcium chloride brine solutions.
3. The contact angles of samples saturated in sodium chloride brine with sodium chloride brine were smaller than the samples saturated in calcium chloride brine with calcium chloride brine.

VIII. DISCLAIMER

REFERENCES

1. BSC 2005, *Technical Work Plan for: Measuring Contact Angle between Brines and Tuff.* Las Vegas, Nevada. ACC. DOC.20050309.0001
2. First Ten Angstroms, 2002, User Manual Drop Shape Analysis, Software Version 2.0, 465 Dinwiddie Street, Portsmouth, Virginia 23704. BSC TIC#256953.
3. BSC 2005, *Data Transmittal: Contact Angle Measurements for Yucca Mountain Tuff with Brines*. ACC:20051201.01212.

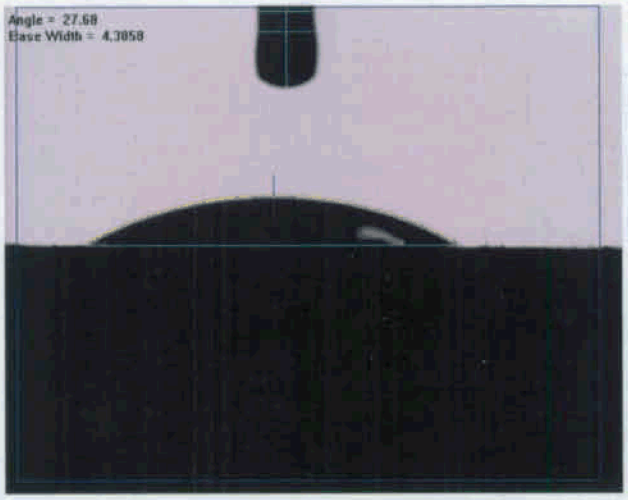

Figure 1A. Calcium Chloride Brine with Calcium Chloride Brine

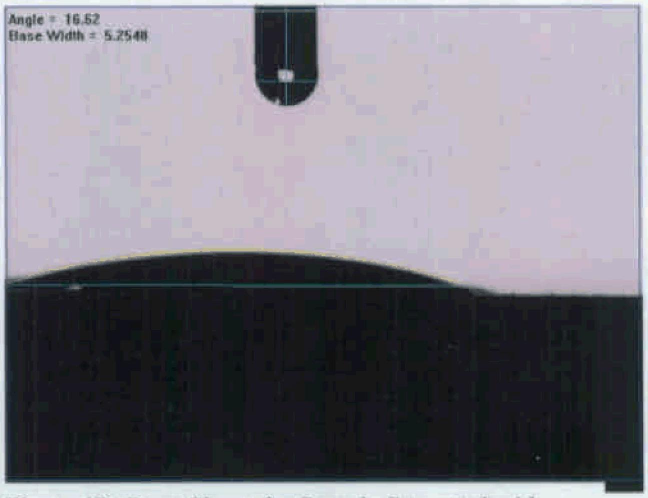

Figure 1B. Drop Shape for Sample Saturated with Sodium Chloride Brine with Sodium Chloride Brine

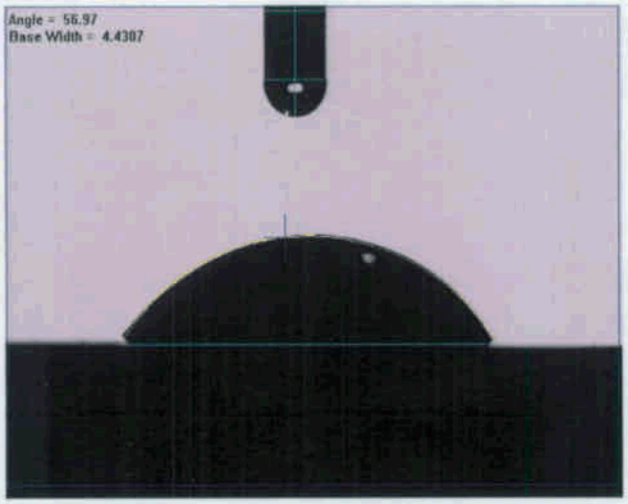

Figure 1C Drop Shape for Sample Saturated in Distilled Water

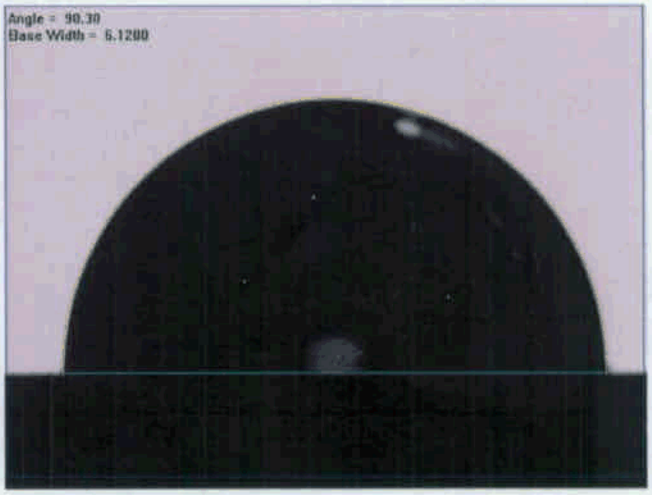

Figure 1D. Shape of Standard 90^0 Angle

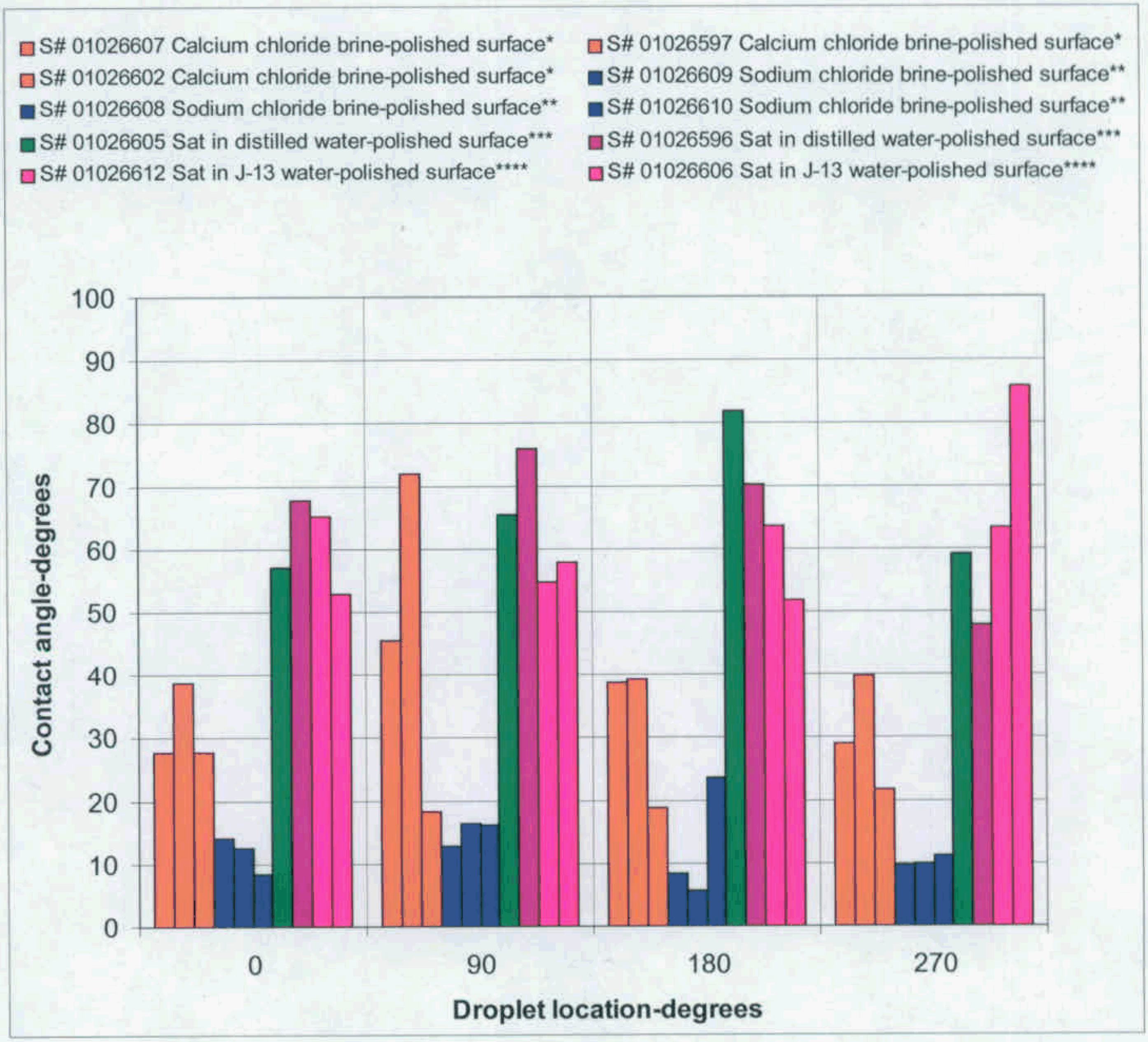

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Figure 2. Contact angle vs. droplet Location for all saturated polished samples

Notes:

* Contact angle for sample saturated in calcium chloride brine with calcium chloride brine

** Contact angle for sample saturated in sodium chloride brine with sodium chloride brine

** Contact angle for sample saturated in distilled water with distilled water

**** Contact angle for sample saturated in J-13 water with J-13 water

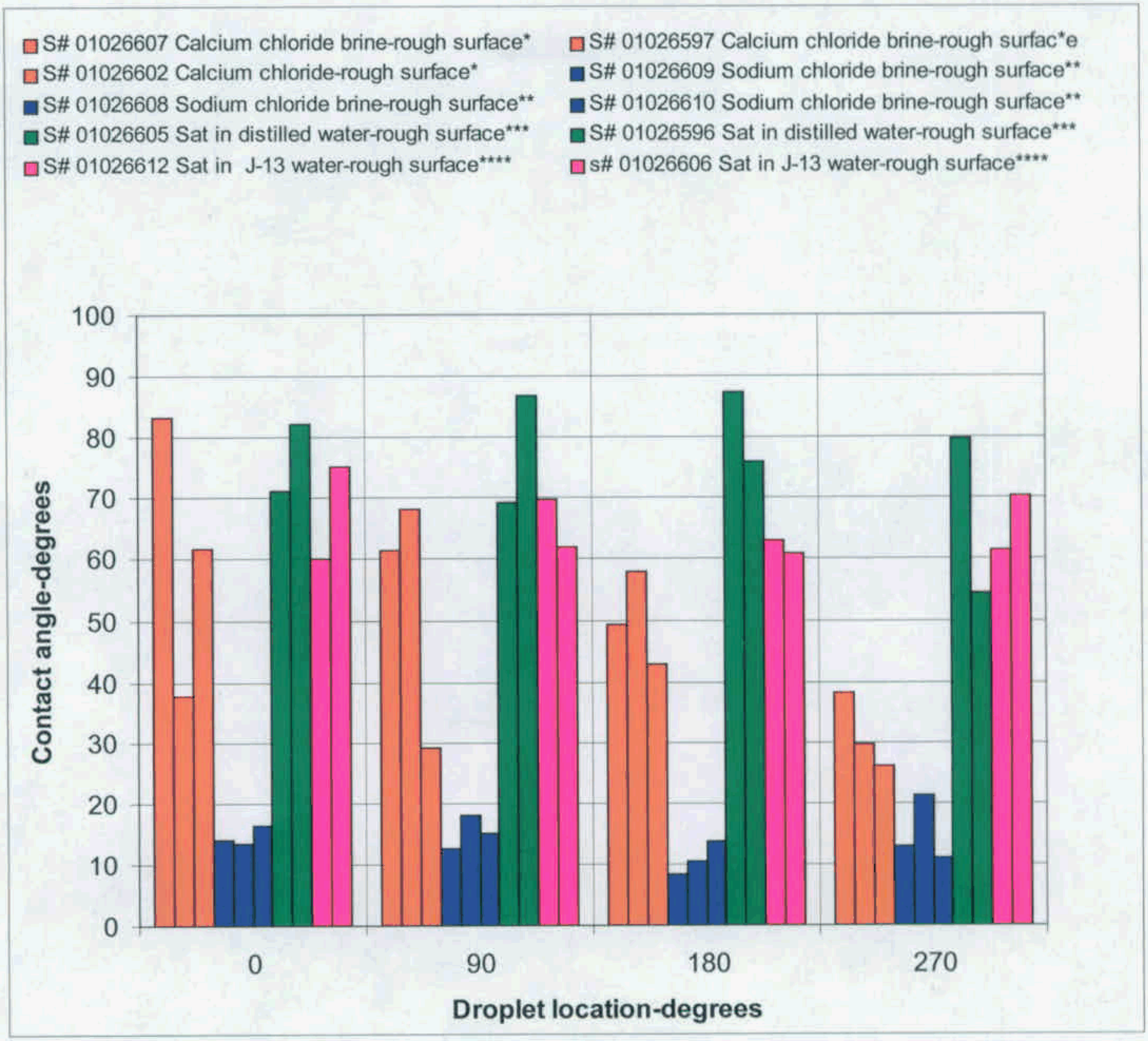

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Figure 3. Contact angle vs. droplet location for all saturated rough samples

Notes:

*	Contact angle for sample saturated in calcium chloride brine with calcium chloride brine
**	Contact angle for sample saturated in sodium chloride brine with sodium chloride brine
**	Contact angle for sample saturated in distilled water with distilled water
****	Contact angle for sample saturated in J-13 water with J-13 water

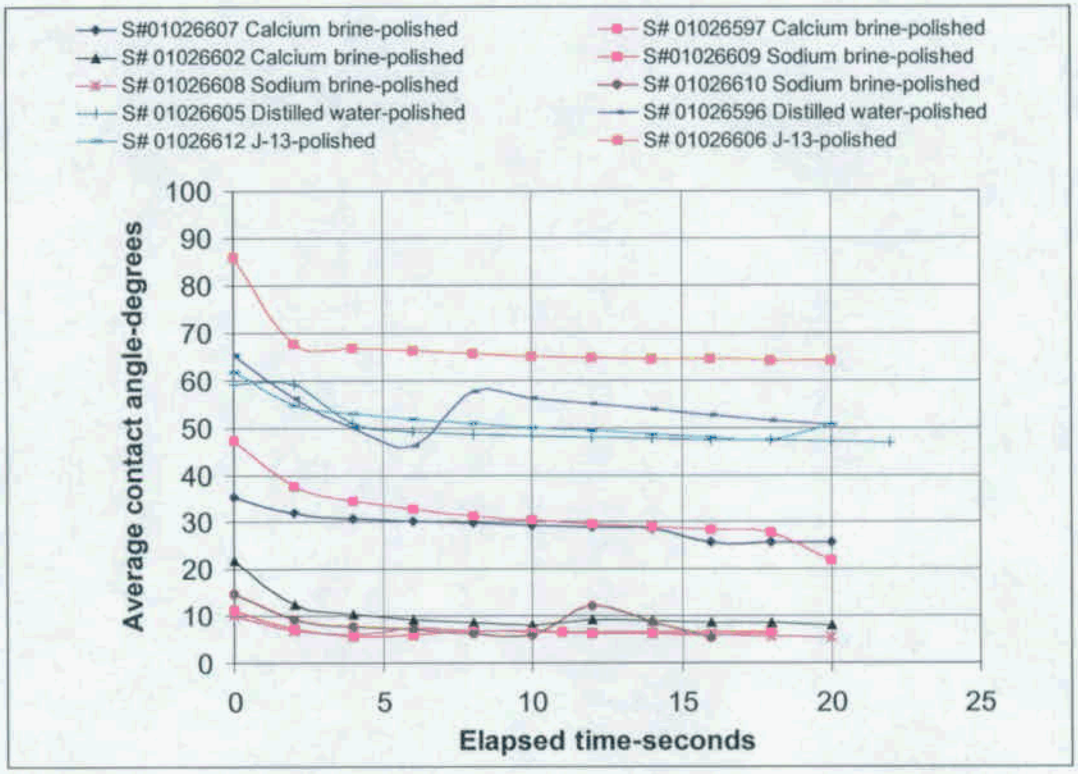

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Figure 4. Average contact angle for saturated samples for polished surface

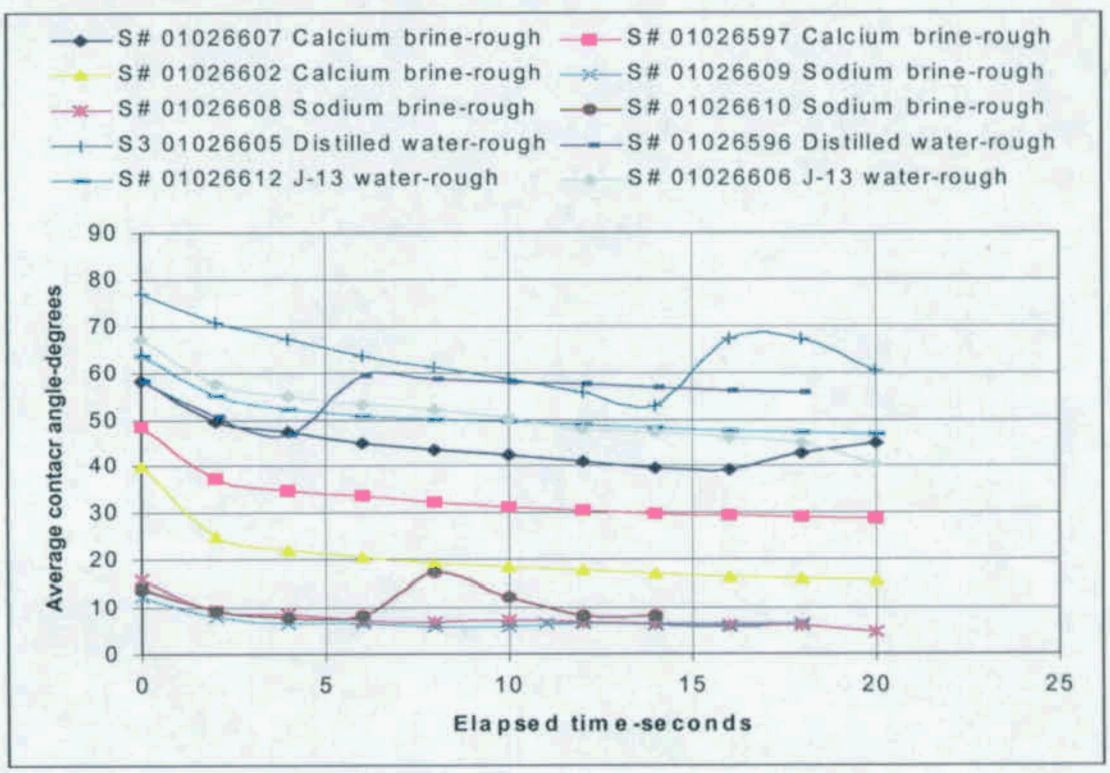

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Figure 5. Average contact angle for saturated samples rough surface